

Nanoparticle Emissions of DI Gasoline Cars with/without GPF

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Abstract

In the present paper some results of investigations of nanoparticles from five DI gasoline cars are represented. The measurements were performed at vehicle tailpipe and in CVS-tunnel. Moreover, five variants of “vehicle – GPF” were investigated. These results originate from the project GasOMeP (Gasoline Organic & Metal Particulates), which focused on metal-nanoparticles (including sub 20nm) from gasoline cars with different engine technologies.

The PN-emission level of the investigated GDI cars in WLTC without GPF is in the same range of magnitude very near to the actual limit value of 6.0×10^{12} #/km. With the GPF's with better filtration quality, it is possible to lower the emissions below the future limit value of 6.0×10^{11} #/km.

There is no visible nuclei mode and the ultrafine particle concentrations below 10nm are insignificant.

Some of the vehicles show at constant speed operation a periodical fluctuation of the NP-emissions, as an effect of the electronic control.

Introduction

The nanoparticles (NP) *) count concentrations are limited in EU for Diesel passenger cars since 2013 and for gasoline cars with direct injection (GDI) since 2014. The limit for GDI was temporary extended to 6×10^{12} #/km (regulation No. 459/2012/EU).

Nuclei of metals as well as organics are suspected to significantly contribute especially to the ultrafine particle size fractions, and thus to the particle number concentration.

The invisible nanoparticles (NP) from combustion processes penetrate easily into the human body through the respiratory and olfactory pathways and carry numerous harmful health effects potentials. The nanoaerosol in vehicle exhaust is known to be a complex mixture of different volatile and non-volatile species often showing a bimodal particle size distribution with a nucleation mode smaller than 20 nm and a larger accumulation mode that mainly contains aggregates of primary particles.

The larger accumulation mode is usually composed of more graphitic soot particles with an elemental carbon (EC) structure, whereas the particles in the nucleation mode are reported to be mainly volatile organics, especially when sulphur is absent from fuel and lubrication oil, [1-4]. However, recent studies detected also low-volatility particle fractions in the ultrafine size range when sampling was carried out according to PMP protocol at 300 °C, [5-7].

These particles are suspected to be nucleated metal oxides originating from metal additives in lubrication oil or fuels [8-11]. The formation of this particulate fraction was especially observed when the soot content was low as in idle condition of diesel vehicles. These particles mainly appear in the ultrafine size range <23 nm. While the mass contribution of these ultrafine particles in vehicle emissions is very low, their contribution to the number concentration is significant. Moreover, these ultrafine particles may contribute to the surface composition of the aerosol and have therefore a significant impact on health effects associated with pollution.

Knowledge about the emission level, chemistry and formation mechanisms of these particles is an important objective in order to assess their toxic potential, and to propose effective measures to reduce these emissions.

Studies for gasoline fuelled internal combustion engines pointed out that also this vehicle class can emit remarkable amounts of particles, [6, 12, 13]. Especially gasoline direct injection technology (GDI) shows particle number (PN) emissions significantly higher than modern diesel cars equipped with best available DPF technology. Since the trend for gasoline vehicles with GDI technology is increasing, a significant rise in emission is predicted in the near future.

The nanoparticles emissions are produced especially at cold start and warm-up conditions and at a dynamic engine operation, [14]. The lube oil contributes to this emission in the sense of number concentrations in nuclei mode and composition, [8, 9, 10].

*) Abbreviations see at the end of this paper

The investigations of morphology of the nanoparticles from gasoline direct injection engine revealed principally graphitic structures, which can store some metal oxides in certain conditions and can be overlapped by condensates, [15, 16].

Car manufacturers and suppliers of exhaust aftertreatment technology offer several mature solutions of GPF for efficient elimination of the nanoparticles from DI SI-engines, [17, 18].

The investigations in present paper were performed at AFHB (Laboratories for IC-Engines and Exhaust Emission Control of the Berne University of Applied Sciences, Biel CH) as a part of the network project GasOMeP, together with the Swiss Research Institutions: EMPA, FHNW and PSI.

Comparisons of NP-emissions of five GDI vehicles at steady state (SMPS) and at transient (CPC) operation, as well as the emissions reduction potentials with different gasoline particle filters (GPF's), are presented.

Tested vehicles

Table 1 summarizes the most important vehicle data. As a reference of the best available technology, concerning the reduction or elimination of PM- and PN-emissions a modern Diesel passenger car with a high-quality DPF was included in the tests (vehicle 6).

Vehicles ①②③	Volvo V60 T4F ①	Opel Insignia 1.6 EcoFlex ②	Mitsubishi Carisma 1.8 GDI ③
Number and arrangement of cylinders	4 / in line	4 / in line	4 / in line
Displacement cm ³	1596	1598	1834
Power kW	132 @ 5700 rpm	125 @ 6000 rpm	90 @ 5500 rpm
Torque Nm	240 @ 1600 rpm	260 @ 1650-3200 rpm	174 @ 3750 rpm
Injection type	DI	DI	DI
Curb weight kg	1554	1701	1315
Gross vehicle weight kg	2110	2120	1750
Drive wheel	Front-wheel drive	Front-wheel drive	Front-wheel drive
Gearbox	a6	m6	m5
First registration	27.01.2012	2014	05.2001
Exhaust	EURO 5a	EURO 5b+	EURO 3
Aftertreatment	TWC	TWC	TWC/Ox.Cat

Table 1a. Data of investigated cars

Vehicles ④⑤⑥	Opel Zafira Tourer ④	VW Golf Plus ⑤	Diesel Peugeot 4008 1.6HdI STT ⑥
Number and arrangement of cylinders	4 / in line	4 / in line	4 / in line
Displacement cm ³	1598	1390	1560
Power kW	125 @ 6000 rpm	118 @ 5800 rpm	84 @ 3600 rpm
Torque Nm	260 @ 1650 - 3200 rpm	240 @ 1500 rpm	270 @ 1750 rpm
Injection type	DI	DI	DI
Curb weight kg	1678	1348 - 1362	1462
Gross vehicle weight kg	2360	1960 - 1980	2060
Drive wheel	Front-wheel drive	Front-wheel drive	Front-wheel drive
Gearbox	m6	m6	m6
First registration	22.07.2014	01.02.2010	12.04.2013
Exhaust	EURO 5b+	EURO 4	EURO 5b
Aftertreatment	TWC	TWC	DPF

Table 1b. Data of investigated cars

Property	Unit	Result
Density (at 15°C)	kg/m ³	736.1
Vapor pressure (at 37.8°C)	kPa	67.3
Research Octan Number (RON)	-	95.6
Oxygen content	% (m/m)	1.0
Sulfur content	mg/kg	<1.0
Pb Lead	mg/L	<1.0
Ca Calcium	mg/kg	<1.0
Fe Iron	mg/kg	<1.0
Mg Magnesium	mg/kg	<1.0
Mn Manganese	mg/kg	<1.0
P Phosphorus	mg/kg	<1.0
Zn Zink	mg/kg	<1.0
Na Natrium	mg/kg	<1.0
K Potasium	mg/kg	<1.0
Distillation (at 101.3 kPa)		
• start	°C	34
• 10% Vol	°C	48
• 50% Vol	°C	75
• 90% Vol	°C	142
• end	°C	174

Table 2. Data of gasoline

The gasoline used was from the Swiss market, RON 95, according to SN EN228. A bigger charge of gasoline was purchased for the project and it was analyzed at INTERTEC Laboratory. The most important data are given in Table 2. The lube oils were also analyzed at EMPA Laboratory, Table 3, which shows the 9 most prominent metals and the sums of all analyzed 21 metals. For all GDI-vehicles, except of vehicle 2, the same lube oil was applied. For the Diesel car the lube oil was not changed and not analyzed.

Property (typical value)	Vehicles ①③④⑤	Vehicle ②	Unit
	Castrol Magnatec	dexos 2	
Viscosity kin 40°C	72.0	72.0	mm ² /s
Viscosity kin 100°C	12.2	12.1	mm ² /s
Viscosity index	166	165	(--)
Density 15°C	852	854	kg/m ³
Pour point	-39	-36	°C
Flash point (PMCC)	207	>201	°C
Total Base Number TBN		7.5	mg KOH/g
Sulphated ash	0.8	0.8	%wt
Na *	4.7	434	µg/g
Mg *	17	9.2	µg/g
Al *	32	5.4	µg/g
Ca *	2240	2300	µg/g
Mn *	0.20	24	µg/g
Fe *	17	34	µg/g
Cu *	0.07	27	µg/g
Zn *	760	630	µg/g
Mo *	36	0.81	µg/g
Sum metals	3109.56	3481.48	µg/g

Table 3. Data of the utilized lube oils (* analysis, others: specifications)

Test methods and instrumentation

The vehicles were tested on a chassis dynamometer at constant speeds and in the dynamic driving cycles WLTC, with cold & warm engine start.

Chassis dynamometer - following test systems were used:

- roller dynamometer: Schenk 500 GS 60
- driver conductor system: Tornado, version 3.3.
- CVS dilution system: Horiba CVS-9500T with Roots blower
- air conditioning in the hall automatic (intake- and dilution air).

The driving resistances of the test bench were set according to the legal prescriptions, responding to the horizontal road.

Nanoparticle analysis

The measurements of NP size distributions were conducted with different SMPS-systems, which enabled different ranges of size analysis:

SMPS: DMA TSI 3081 & CPC TSI 3772 (10 - 429 nm)
nSMPS: nDMA TSI 3085 & CPC TSI 3776 (2 - 64 nm)

For the dilution and sample preparation an ASET system from Matter Aerosol was used, (ASET ... aerosol sampling & evaporation tube). This system contains:

- Primary dilution - MD19 tunable rotating disc minidiluter (Matter Eng. MD19-2E)
- Secondary dilution – dilution of the primary diluted and thermally conditioned sample gas on the outlet of evaporative tube.
- Thermoconditioner (TC) - sample heating at 300°C

This sample preparation system fulfills the requirements of PMP and it was used for all measurements. At steady state operation (SSC see next section) this system worked with summary dilution factors DF = 100 to 500.

The estimated accuracy of PN-measurement in the size range of 80-120 nm, with DF = 100 is +/- 6%.

In the tests the gas sample for the NP-analysis was taken from the undiluted exhaust gas at tailpipe for stationary operation (SMPS) or from the diluted exhaust gas in CVS-tunnel at transient operation (CPC). The schematic of the general sampling set up is represented in Fig. 1.

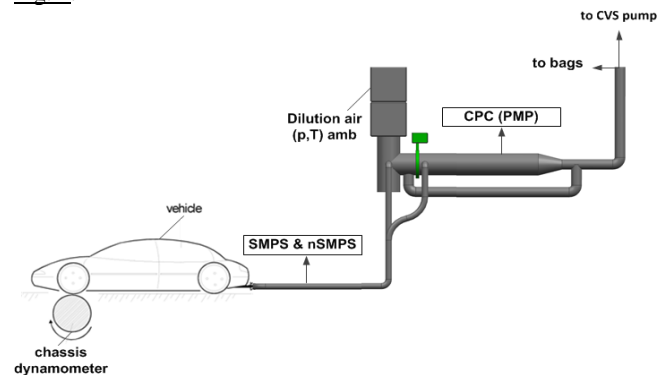


Fig. 1 Set-up of exhaust gas sampling for PN-analysis

Driving cycles

The vehicles were tested on a chassis dynamometer at constant speeds (SSC) and in the dynamic driving cycles (WLTC).

The steady state cycle (SSC) consists of 20 min-steps at 95, 61, 45, 26 km/h and idling, performed in the sequence from the highest to the lowest speed.

Fig. 2 shows the steady state cycle (SSC) with the resulting tailpipe temperatures (t_{exh}) for gasoline vehicle 1. This gives the magnitude of the temperatures at the particulate sampling point “tailpipe” during steady state measurements (SMPS).

The approach to find a homogenized world-wide driving cycle was successfully finished with the development of the homogenized WLTP world-wide light duty test procedure. The WLTC (world-wide light duty test cycle) represents typical driving conditions around the world.

This cycle (Fig. 3) has been used also in this study. It represents different driving conditions: urban, rural, highway and extra-highway.

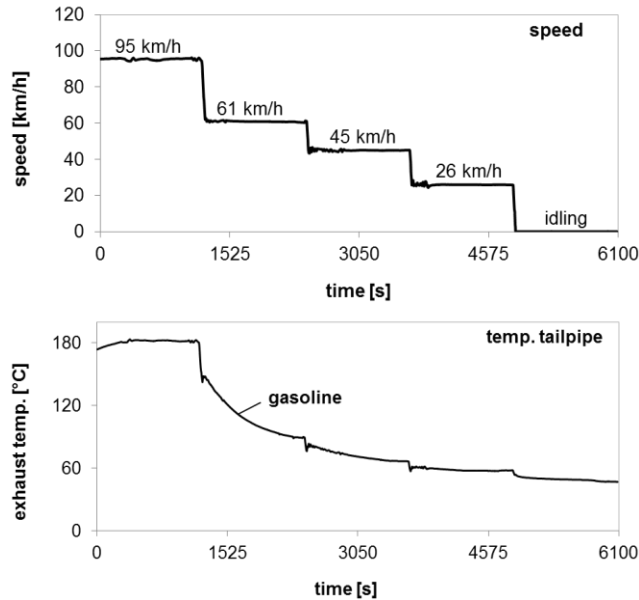


Figure 2. SSC steady state cycle and tailpipe temperature of vehicle 1

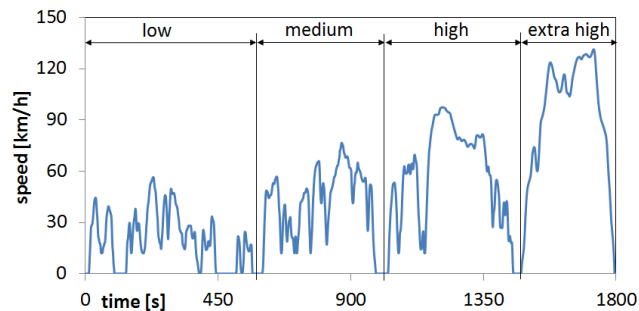


Figure 3. WLTC driving cycle

Results

Steady state operation (SSC)

The considerations of particle size distributions at steady state operation give a basic view on the PN-concentrations at tailpipe and allow some reflections about the nanoparticle production. Nevertheless, this is not a legal measuring procedure and therefore the results does not have to be compared with the legal PN limit values.

Fig. 4 represents exemplary the SMPS particle size distributions (PSD) of all tested vehicles (V1 to V5) at tailpipe without GPF at the same constant speeds and idling.

At 95 and 45 km/h the maxima of PSD's show in certain cases the particle counts concentrations (PC) in the range of 10^6 to 10^7 #/cm³, which is similar as for Diesel engines (without DPF). At idling, the PC values are roughly one order of magnitude lower.

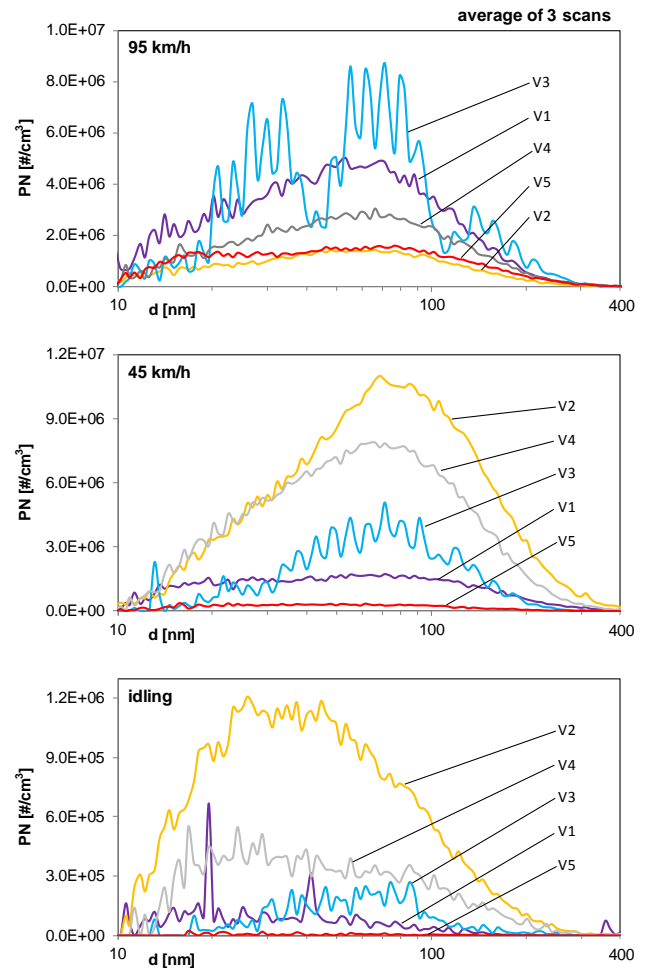


Figure 4. SMPS particle size distributions at constant speeds with different GDI vehicles (w/o GPF).

For vehicle 3, strong fluctuations of the PC-concentration during the period of scanning (over the size range) are visible. During the constant speed operation of this vehicle (at 95 and 45 km/h), periodic fluctuations of gaseous emissions (CO, HC, NO_x) were observed (not represented here) and confirmed a continuous switching of the operation between lean and rich. This means that for this vehicle, changing between the stratified, or homogenous (lean) and homogenous (rich) operating strategies, it also implies the switching of parameters, like ignition timing, injection timing, injection quantity and eventually EGR. This can have the influences on NP-emissions as demonstrated.

The relationships of NP-emissions between different vehicles can vary depending on operating condition. As example: vehicle 2 has at 95 km/h the lowest and at 45 km/h and idling the highest particle counts concentrations. This is also visible in the summary representation of integral PN-emissions at all tested constant speeds, Fig. 5.

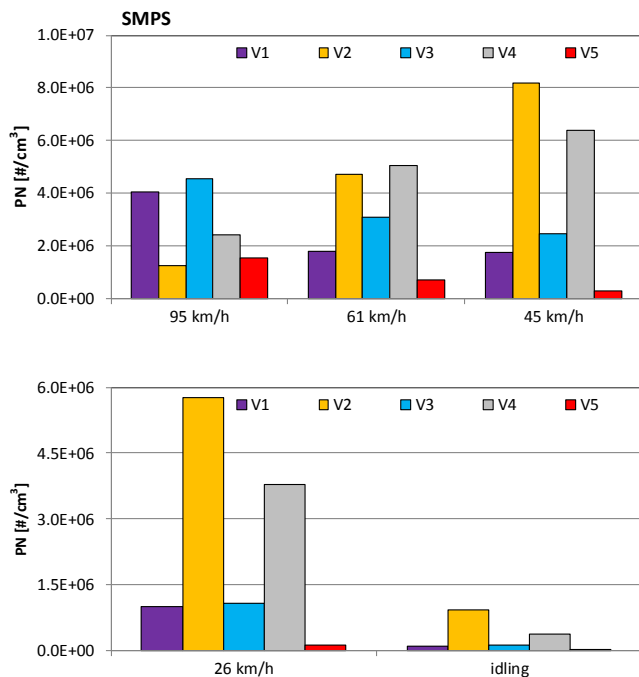


Figure 5. Integral PN emissions at constant speeds with different GDI vehicles (w/o GPF).

There are different interacting processes during mixture preparation, combustion and gas flow in the exhaust system, which sensitively influence the generation of nanoparticle emissions. The following discussion gives some ideas and hypotheses about the reasons of the observed differences of PN-results between the different vehicles:

Important question is the mixture preparation: the ideal mixture preparation should atomize and evaporate all the used fuel and bring it as homogeneously premixed, as possible into the combustion chamber.

For MPI there is usually a portion of fuel deposited on the walls of the intake port, which can, especially at transient operation, arrive in the combustion chamber as liquid non-premixed droplets. A part of this “unprepared” fuel burns heterogeneously and is a source of soot-production.

These effects are stronger in DI technology and especially, when the liquid fuel arrives at the wall and, what is also possible, interacts with the lube oil layer, the production of nanoparticles is particularly increased, [19, 20].

The chemistry of oil and fuel, their HC-matrix and additive packages have a significant influence on the NP’s.

Further to consider are: the passage of aerosol through the exhaust system, the history of temperature drop, catalysis, chemistry, spontaneous condensation and store/release effects in the exhaust system. All of them have finally influences on “what will be measured at tailpipe”.

The processes influencing NP-production depend on engine operating conditions. With no doubt the NP-emissions vary with the operating point and are increased at transient operation.

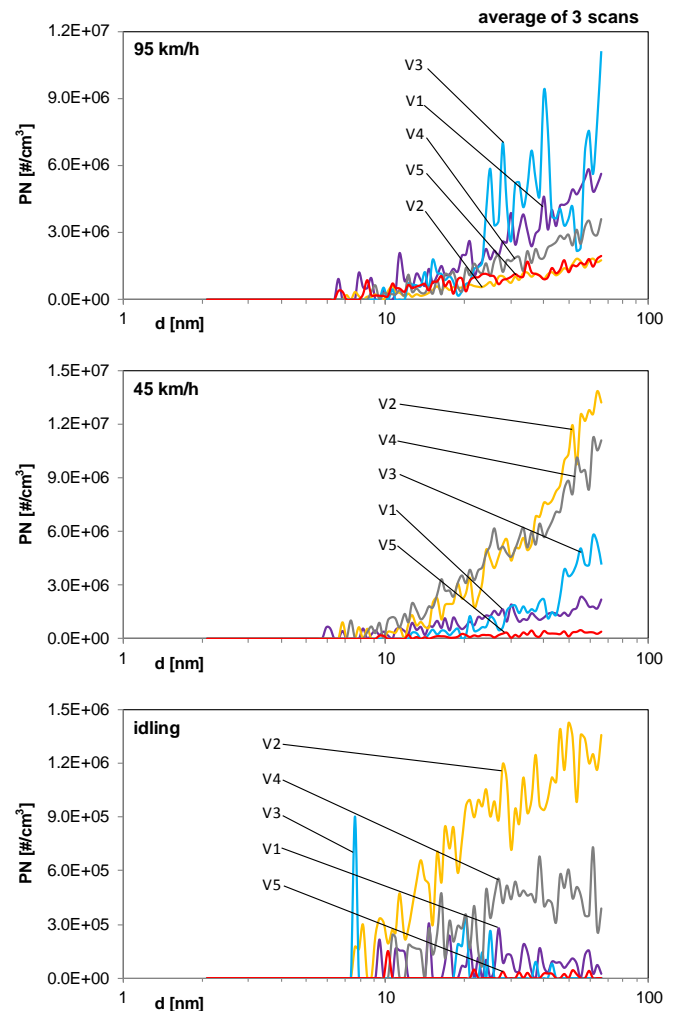


Figure 6. nSMPS particle size distributions at constant speeds with different GDI vehicles (w/o GPF).

The measurements of all PSD’s at constant speeds were simultaneously performed with two systems SMPS (size range 10-429 nm) and nano-SMPS (size range 2-64 nm). Fig. 6 shows the results obtained with nSMPS with all vehicles (w/o GPF) at the three chosen constant operating conditions. It can be remarked, that the NP-concentrations with different vehicles show the same tendencies as in Fig. 4. There are no PC in the sizes below 6 nm and the PC in the size range 6 to 10 nm can be considered as negligible.

Generally there is a very good accordance of PSD’s measured with both systems SMPS and nSMPS in the common size range (10-64 nm). Fig. 7 shows an example of scans with and without GPF. It confirms the excellent accordance of scans with both systems, it also confirms a very good particle count filtration efficiency (PCFE) of the tested GPF and it particularly shows the total elimination of nanoparticles with sizes below 30 nm.

The opinion of the authors, resulting from these tests as well as from previous experiences with GDI-vehicles, [21], is that additional research, or discussions about NP’s with sub-10 nm-sizes and more restrictions of the legislation for sub-23nm-sizes, are not necessary.

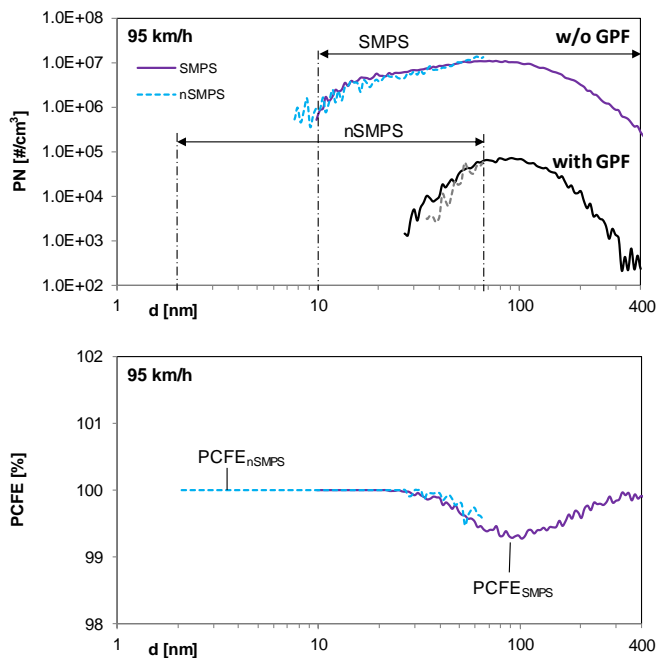


Figure 7. Example of PSD's with SMPS & nSMPS and particle counts filtration efficiency (PCFE) with V1, GPF 1 at 95 km/h

Five variants “vehicle – GPF” were tested. The GPF’s were randomly obtained for the tests and they were mounted in the exhaust systems of the cars approximately 60 cm downstream of the TWC. They were neither developed, nor optimized for this application. The specific data of the GPF’s are not available.

Fig. 8 summarizes the filtration efficiencies (PCFE) obtained at the constant speeds. The PCFE-values are between 91% and 100%. GPF3 and GPF4 represent clearly lower filtration efficiency than GPF1 and GPF2. This result indicates that the filtration efficiencies can be adapted by optimizing the substrate to fulfil different objectives or requirements.

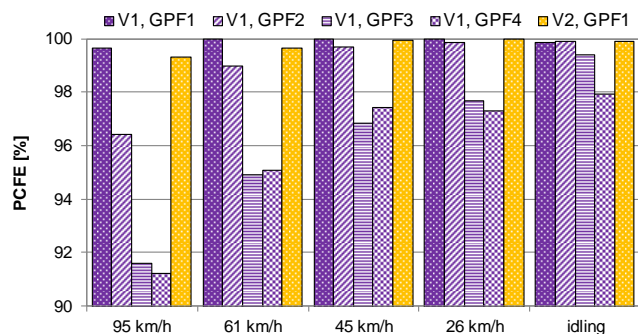


Figure 8. Filtration efficiencies PCFE at constant speeds with different GPF's (SMPS data).

In comparison, the quality requirements for DPF retrofitting are: for the Swiss Confederation OAPC $PCFE \geq 97\%$ and of the VERT Association $PCFE \geq 99\%$. This is in the sense of “best available technology for health protection”.

Transient operation

The results at transient operation are obtained with CPC (according to PMP) at the end of CVS-dilution tunnel. These results can be compared with the legal PN limit values.

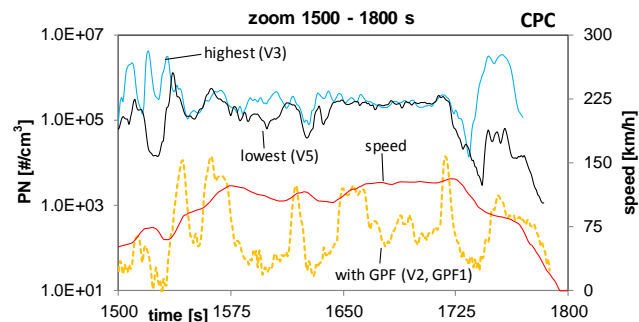


Figure 9. Examples of PN time-courses with different vehicles in the high-speed part of WLTC hot.

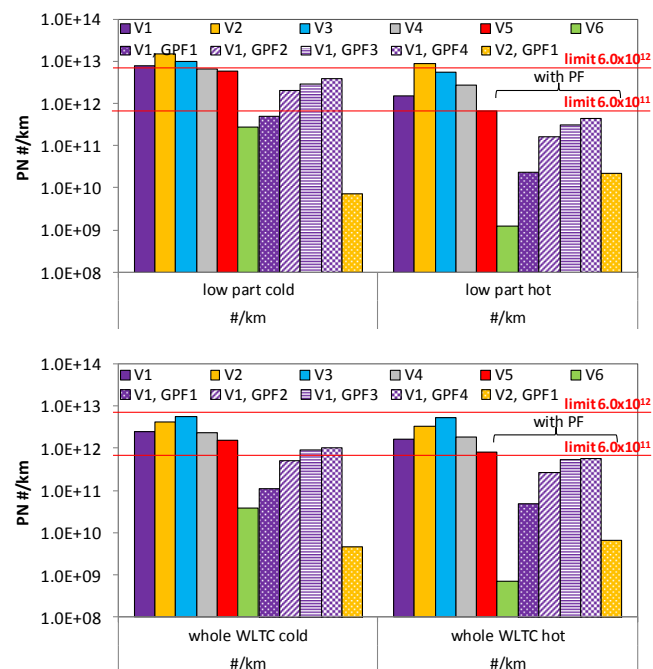


Figure 10. Comparison of PN-emissions in WLTC cold and hot for different vehicles

All mechanisms influencing the NP-production in combustion chambers and in the exhaust system are at transient operation variable and mostly overlapping each other. A known and accepted fact is that the peak values of NP-emissions coincide with the acceleration, or deceleration events in the driving cycle. Fig. 9 shows an example of time-plots of NP-emissions with high- and low emitting vehicles in the high-speed part of WLTC. There are the highest peak-values at the strongest acceleration, or deceleration. The PN-concentrations with GPF have very low absolute values (below the ambient background) and the natural fluctuations of emissions overlap the results and are more predominant than the effects of the driving cycle.

Fig. 10 summarizes the average PN emissions in WLTC cold and hot. The emission level of “hot” cycles is generally lower than the emission level of “cold” cycles. Vehicles which are equipped with GPF have, as expected, lower PN-emissions. Vehicle 6 is a Diesel car with original DPF of a very good quality; it sets a quality level, which is only roughly attained by the vehicle 2 with GPF1.

From all variants with GPF's the GPF3 and GPF4 have the highest emissions. These two filters also have the lowest average filtration efficiencies, Fig. 11.

Finally, it can be concluded that the PN-emission level of the investigated GDI cars in WLTC without GPF is in the same range of magnitude very near to the actual limit value of 6.0×10^{12} #/km. With the GPF's with better filtration quality it is possible to lower the emissions below the future limit value of 6.0×10^{11} #/km.

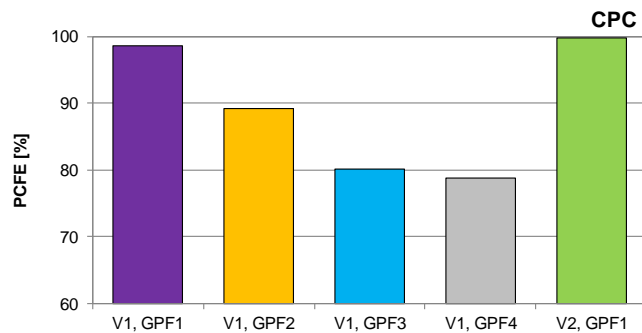


Figure 11. PCFE's of the investigated GPF's in WLTC hot

Conclusions

The most important statements of this work can be summarized as follows:

- The PN-emission level of the investigated GDI cars in WLTC without GPF is in the same range of magnitude very near to the actual limit value of 6.0×10^{12} #/km.
- With the GPF's with better filtration quality it is possible to lower the emissions below the future limit value of 6.0×10^{11} #/km.
- The filtration efficiency of GPF can attain 99% but it can also be optimized to lower values – in this respect the requirement of “best available technology for health protection” should be considered.
- The relationships of NP-emissions between different vehicles can vary depending on operating condition.
- Generally there is a very good accordance of PSD's measured with both systems SMPS and nSMPS in the common size range (10-64 nm).
- For the vehicles with gasoline DI, there is no increase of PC's in nuclei mode (below 10 nm) at the measured constant speeds, the particle counts below 10 nm are negligible.
- Due to the electronic regulation of the engine the NP-emission of some vehicles (here vehicle 3) are periodically fluctuating.
- There is a good repeatability of the average emissions in the “warm” driving cycles.
- Comparing the NP-emissions of different vehicles with SMPS PSD's at constant operation gives only a limited information about the relationships of emissions measured with CPC in dynamic driving cycles.

The present paper focuses solely on solid nanoparticle emissions. The tested gasoline cars, except of vehicle 3, were with homogenous combustion concept and represented a modern TWC technology. According to that the emissions of gaseous legislated components (CO, HC, NO_x) were very low.

Research on an older MPI vehicle, [21], showed tendencies of significantly increased PN-emissions.

The present high filtration quality of Diesel vehicles (DPF) set's high requirements on the filtration quality in the gasoline sector (GPF).

Acknowledgements

The authors want to express their gratitude to the institutions, which financially supported the activities: Swiss Federal Office of Environment, Swiss Federal Office of Energy, Swiss Oil and Swiss Lubes.

For technical discussions, inspirations and help thanks are due to the GasOMeP partners: Dr. Norbert Heeb, EMPA; Prof. Dr. Heinz Burtcher, FHNW and Dr. André Prévot, PSI.

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Definitions/Abbreviations

AFHB	Abgasprüfstelle FH Biel, CH
ASET	Aerosol Sampling & Evaporation Tube
BAFU	Bundesamt für Umwelt, (see FOEN)
CLA	chemiluminescent analyzer
CPC	condensation particle counter

CVS	constant volume sampling
DF	dilution factor
DI	Direct Injection
DMA	differential mobility analyzer
DPF	Diesel particle filter
EC	Elemental Carbon European Community
EGR	exhaust gas recirculation
EMPA	Eidgenössische Material Prüf- und Forschungsanstalt
FHNW	Fachhochschule Nord-West Schweiz
FOEN	Federal Office for Environment
GasOMeP	Gasoline Organic & Metal Particles
GDI	gasoline direct injection
GPF	gasoline particle filter
GRPE	EC Groupe Rapporteurs Pollution & Energy
MD	minidiluter
MFS	mass flow sensor
MPI	multipoint port injection
NP	nanoparticles < 999 nm
nSMPS	nano SMPS
OAPC	CH: Ordinance of Air Protection Control
PC	particle counts (integrated)
PM	particle mass
PN	particle numbers
PMP	Particle Measuring Program of the GRPE
PSD	particle size distribution
PSI	Paul Scherrer Institute

SMPS	scanning mobility particle sizer	V	vehicle
TC	thermoconditioner	VERT	Verification of Emission Reduction Technologies
TTM	Technik Thermische Maschinen	WLTC	Worldwide Light Duty Test Cycle
TWC	three way catalyst		